

Oscillator Strengths and Predissociation Rates for Rydberg Transitions in $^{12}\text{C}^{16}\text{O}$, $^{13}\text{C}^{16}\text{O}$, and $^{13}\text{C}^{18}\text{O}$ Involving the $E\ ^1\Pi$, $B\ ^1\Sigma^+$, and $W\ ^1\Pi$ States

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ABSTRACT

One of the processes controlling the interstellar CO abundance and the ratio of its isotopologues is photodissociation. Accurate oscillator strengths and predissociation rates for Rydberg transitions are needed for modeling this process. We present results on absorption from the $E\ ^1\Pi - X\ ^1\Sigma^+$ (1-0) and $B\ ^1\Sigma^+ - X\ ^1\Sigma^+$ (6-0) bands at 1051 and 1002 Å, respectively, and the vibrational progression $W\ ^1\Pi - X\ ^1\Sigma^+$ ($v'-0$) bands with $v' = 0$ to 3 at 972, 956, 941, and 925 Å, respectively. The corresponding spectra were acquired at the high resolution ($R \approx 30,000$) SU5 beam line at the Super ACO Synchrotron in Orsay, France. Spectra were obtained for the $^{12}\text{C}^{16}\text{O}$, $^{13}\text{C}^{16}\text{O}$, and $^{13}\text{C}^{18}\text{O}$ isotopologues. These represent the most complete set of measurements available. Comparison is made with earlier results, both empirical and theoretical. While earlier determinations of oscillator strengths based on absorption from synchrotron radiation tend to be somewhat smaller than ours, the suite of measurements from a variety of techniques agree for the most part considering the mutual uncertainties. For the bands studied here, their relative weakness, or their significant line widths arising from predissociation, minimizes potential problems from large optical depths at line center in absorption measurements. Predissociating line widths could generally be extracted from the spectra thanks to the profile simulations used in the analysis. In many cases, these simulations allowed us to consider e and f parity levels separately and to determine the dependence of the width on rotational quantum number, J . Our results are consistent with earlier determinations, especially the widths inferred from laser experiments.

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1. Introduction

The photochemistry of CO plays an important role in photon dominated regions (PDRs) of interstellar clouds, circumstellar shells surrounding AGB stars, planetary nebulae, circumstellar disks around young stars, and comets. Models of these regions (e.g., van Dishoeck & Black 1988; Sternberg & Dalgarno 1995; Hollenbach & Tielens 1997, and references therein) use observational data on CO transitions to extract physical conditions. The models rely on oscillator strengths for dissociating transitions (e.g., Letzelter et al. 1987; Eidelsberg et al. 1991; Chan, Cooper, & Brion 1993; Federman et al. 2001; Sheffer, Federman, & Andersson 2003; Eidelsberg et al. 2004a). While a consensus is emerging for oscillator strengths pertaining to many of the important electronic transitions (Federman et al. 2001; Eidelsberg et al. 2004a), the large number of transitions leading to photodissociation warrants a careful assessment of as many measurements as possible. Here we present results on the $E\ ^1\Pi - X\ ^1\Sigma^+$ (1-0), $B\ ^1\Sigma^+ - X\ ^1\Sigma^+$ (6-0), and core excited $W(A^2\Pi^+) - X\ ^1\Sigma^+$ ($v'-0$) bands with $v' = 0$ to 3 for $^{12}\text{C}^{16}\text{O}$, $^{13}\text{C}^{16}\text{O}$, and $^{13}\text{C}^{18}\text{O}$ acquired at the Super ACO synchrotron source in Orsay, France.

These results are a continuation of our earlier efforts. Federman et al. (2001) studied Rydberg transitions from the ground vibrational level in $^{12}\text{C}^{16}\text{O}$ between 1150 Å and about 1075 Å. The limitation on the short wavelength end was the presence of LiF windows for the gas cell. More recently, a system without windows based on differential pumping allowed us to measure CO absorption from bands with wavelengths approaching 912 Å, the Lyman limit. Eidelsberg et al. (2004a) analyzed the strongly interacting bands [$K - X$ (0-0), $L' - X$ (1-0), and $L - X$ (0-0)] near 970 Å in $^{12}\text{C}^{16}\text{O}$, $^{13}\text{C}^{16}\text{O}$, and $^{13}\text{C}^{18}\text{O}$. We now turn our attention to the other transitions involving Rydberg states that were acquired during the same synchrotron runs. The transitions discussed in the present paper are seen in interstellar spectra (Sheffer et al. 2003) and are important for CO photodissociation (van Dishoeck & Black 1988).

2. Experimental Details and Data Analysis

Since our spectra were acquired during the runs described by Eidelsberg et al. (2004a), we only provide a brief overview here. The measurements were obtained on the Super ACO

synchrotron ring using the high spectral resolution SU5 beam line (Nahon et al. 2001a, b). The spectrometer consisted of a 2400 lines mm^{-1} SiC grating blazed at 13 eV. The entrance and exit slits were set at 35 or 25 μm , yielding respective instrumental widths of 30 or 20 mÅ, as deduced from the analysis based on profile simulation described below. The vacuum ultraviolet (VUV) light emerged from the spectrometer and passed through a windowless gas cell; differential pumping maintained the ultra-high vacuum in the spectrometer and storage ring. High purity CO gas ($^{12}\text{C}^{16}\text{O}$ – Alphagaz, 99.997%; $^{13}\text{C}^{16}\text{O}$ – Eurisotop, 99.1% ^{13}C , 99.95% ^{16}O ; $^{13}\text{C}^{18}\text{O}$ – Isotech, 98.8% ^{13}C , 94.9% ^{18}O) continuously flowed through the cell. The pressure in the cell, typically 1 to 10 mTorr except for the weakest bands, was measured with a capacitance gauge whose full scale was 10 Torr. The stability of the pressure was checked by monitoring a cold cathode ionization gauge in the differentially pumped section where typical pressures were 10^{-6} Torr. The VUV flux exiting the gas cell was detected with a photomultiplier tube (EMI 9558QB) placed after a sodium salicylate coated window acting as a scintillator. The digitized signal from the PMT was recorded on a PC.

Oscillator strengths (f -values) were derived through syntheses of the measured absorption bands. As noted in Eidelsberg et al. (2004a), two independent codes were used to check for consistency, where agreement at the 2% level was achieved. Previous analyses of absorption from Rydberg bands of CO with the Toledo-based code included the experimental work of Federman et al. (2001) and the interstellar study of Sheffer et al. (2003). The code developed in Meudon was used on $A - X$ bands (Eidelsberg et al. 1999) and intersystem transitions (Rostas et al. 2000). The synthetic spectra were based on tabulated spectroscopic data (Eidelsberg et al. 1991). Each synthetic spectrum was adjusted to match the experimental one in a non-linear least-squares fitting procedure with band oscillator strength, line width (instrumental, thermal, and predissociation), and wavelength offset as free parameters. Initial fits revealed that the thermal width was consistent with Doppler broadening at 295 K and was held constant in subsequent syntheses. Predissociation widths of 0.34 mÅ for the $E - X$ (1-0) band in $^{12}\text{C}^{16}\text{O}$, 0.27 mÅ in $^{13}\text{C}^{16}\text{O}$, and 0.18 mÅ for $^{13}\text{C}^{18}\text{O}$ were taken from Ubachs, Velchev, and Cacciani (2000). (Hereafter we use the shorthand $E1$ because absorption examined in the present paper always involves the ground vibrational level.) For the $W0$, $W2$, and $W3$ bands, a J -dependent width had to be introduced in the synthetic spectra in order to reproduce the observations, in keeping with the results of Drabbels et al. (1993) and Eikema, Hogervorst, and Ubachs (1994).

The situation for the $W2$ bands required special care. According to Eikema et al. (1994), the predissociation width is independent of J until $J = 6$, but dependent on J for $J > 6$ for e and f components in all isotopologues. We first determined the J -independent part of the width from lines up to Q(6) and P(3), and then fitted the whole band to derive the J -dependent part and the f -value.

The first step in the analysis was to obtain an accurate measure of the CO column in the differentially-pumped system. This was achieved through profile fits of the $E0$ band near 1075 Å for each isotopologue. Since the $3p\pi$ ($v' = 0$) state is relatively isolated and can be considered essentially free of perturbations, the band f -value was assumed to be the same for the three isotopologues. Predissociation widths of 0.067 mÅ for $^{12}\text{C}^{16}\text{O}$, 0.049 mÅ for $^{13}\text{C}^{16}\text{O}$, and 0.047 mÅ for $^{13}\text{C}^{18}\text{O}$ (Cacciani et al. 1998) were used as input. The pressure deduced from the fit to the $E0$ band was typically within 10% of the value given by the pressure gauge. The deduced pressure was then adopted in syntheses of the bands of primary interest scanned with the same set of conditions. For the high pressures needed to measure absorption from the $E1$ band of $^{13}\text{C}^{18}\text{O}$ – 24 to 58 mTorr, the values from the pressure gauge were used.

Examples of our fits for $E1$, $W0$ and $W3$, and $B6$ bands appear in Figs. 1 to 3, respectively. The ordering is based on predissociation width, from narrowest to broadest. These figures reveal that several of the bands experience perturbations. In Fig. 1 accidental predissociation at $J_e = 7$ for $^{13}\text{C}^{16}\text{O}$ (and $^{12}\text{C}^{16}\text{O}$) (Stark et al. 1992; Cacciani, Hogerworst, & Ubachs 1995) gives rise to extra lines appearing with R(6) and P(8). In $^{13}\text{C}^{18}\text{O}$ it appears at $J_e = 1$ and 5 (Ubachs et al. 2000) and gives rise to line shifts and broadening that are barely visible in our spectra. These interactions arise from spin-orbit coupling between $E^1\Pi$ ($v = 1$) and $k^3\Pi$ ($v = 6$). As for Fig. 3, the absorption bands at 1002.56 Å in $^{12}\text{C}^{16}\text{O}$, at 1002.81 Å in $^{13}\text{C}^{16}\text{O}$, and at 1003.21 Å in $^{13}\text{C}^{18}\text{O}$ have been attributed to a transition between the ground state $X^1\Sigma^+$ $v'' = 0$ and a resonant state due to the strong coupling between the $3s\sigma B^1\Sigma^+$ Rydberg state and the repulsive $D'^1\Sigma^+$ valence state (modeled by Tchang-Brillet et al. 1992; Monnerville & Robbe 1994; Li et al. 1997; Andric et al. 2004). This resonant state has been discussed in detail and identified by its Rydberg character to the vibrational level $v' = 6$ of the $B^1\Sigma^+$ state by Eidelsberg et al. (2004b). As predicted by Andric et al. (2004), the broadening of these bands varies widely from one isotopologue to another. Very weak and diffuse absorption bands involving the $v' = 4$ and 5 levels have been observed recently (Baker 2005) below the avoided crossing of the diabatic potential curves. $B^1\Sigma^+$ $v' = 6$ is thus the first resonance state to be observed above the avoided crossing. Two other processes affecting the f -values for $B6$ and $W1$ in $^{13}\text{C}^{18}\text{O}$ are discussed in the next section.

3. Results and Discussion

Our results are presented in Table 1 (f -values) and Table 2 (predissociation rates, k_p) along with values from earlier studies. The (FWHM) Lorentzian line widths (Γ_L) that

were determined from the spectra are linked to the corresponding upper level lifetimes (τ) and transition rates (k) by the well known relation, $\Gamma_L = 1/(2\pi\tau) = (1/2\pi)k$ ($k = \Gamma_L(\text{m}\text{\AA}) [1000/\lambda(\text{\AA})]^2 \text{ s}^{-1}$, where λ is the band origin for the transition). The transition rate k is the sum of the radiative and predissociation parts (k_r and k_p , respectively). In the present experiments, the fact that Γ_L can be measured at all implies that k_p is considerably larger than k_r .

Previous empirical f -values are based on synchrotron measurements (Eidelsberg et al. 1991; Stark et al. 1991, 1992, 1993; Yoshino et al. 1995), electron-energy-loss spectroscopy (Chan et al. 1993; Zhong et al. 1997), and analysis of interstellar spectra (Sheffer et al. 2003). As for the predissociation rates, an order of magnitude was first obtained by Eidelsberg et al. (1991) from widths evaluated from absorption features recorded in low-resolution synchrotron spectra. Subsequent efforts (Levelt, Ubachs, & Hogervorst 1992a,b; Drabbels et al. 1993; Eikema et al. 1994; Komatsu et al. 1995) were based on high-resolution line width measurements using laser techniques.

Table 1 lists the results on oscillator strength. Columns 1-3 give the transition, wavelength of the band origin, and isotopologue, respectively. Our f -values appear in the fourth column, and those from previous studies are shown in the remaining ones. We indicate temperature for the measurements. For the transitions examined here, there generally is no difference in f -values among isotopologues within the uncertainty of our measurements.

One exception involves the $B6$ band of $^{13}\text{C}^{18}\text{O}$, the f -value of which is significantly smaller than that of the other isotopologues. It is to be noted that the $3d\sigma - X^1\Sigma^+$ (0-0) band appears on the blue wing of the $B6$ band in $^{13}\text{C}^{18}\text{O}$, while it is not seen in the other two isotopologues at the pressures used here (< 24 mTorr). As was shown in the detailed study of Eidelsberg et al. (2004b), the $3d\sigma 0$ band borrows its intensity from the $B6$ band. This effect is weak for the first two isotopologues, allowing this band to be seen only at high column density. For the $B6$ band of $^{13}\text{C}^{18}\text{O}$, it appears that the mixing is stronger, leading to similar intensities for the two bands. The pertinent value to consider is thus the sum of the $B6$ and $3d\sigma 0$ oscillator strengths. This is about 0.0066, a value more in line with the f -value for the other isotopologues. The second exception, $W1$ in $^{13}\text{C}^{18}\text{O}$, is so severe that we are not able to derive an f -value. This band is strongly overlapped by another band of unknown origin, whose profile does not resemble a $\pi\sigma$ or $\sigma\sigma$ band. Since its intensity varies with pressure as $W1$ does, the band probably arises from a valence state perturber. Thus it is not possible to separate its contribution from that of $W1$. Finally, we note that the f -values for the W bands are relatively strong, with the (2-0) band being the strongest. The results for the W bands are consistent with the theoretical predictions of Cooper & Kirby (1988) – i.e., f -values of about 10^{-2} and Franck-Condon factors favoring the (2-0) band.

Our oscillator strengths are generally consistent with earlier determinations. However, the previous absorption experiments using synchrotron sources (Eidelsberg et al. 1991; Stark et al. 1991, 1992, 1993; Yoshino et al. 1995) tend to have somewhat smaller f -values than ours and those from the interstellar study of Sheffer et al. (2003), even though the results agree within their mutual uncertainties. Potential problems with optically thick absorption noted by Federman et al. (2001), Sheffer et al. (2003), and Eidelsberg et al. (2004a) are less of an issue for the current set of transitions because they are weaker or have intrinsic line widths that are broadened by predissociation (see below). The one instance where optical depth effects may have played a role involves the results for $E1$ of Eidelsberg et al. (1991) since predissociation is a minor channel for this band.

A comparison of predissociation rates appears in Table 2. Whenever possible, results for e and f parity are given separately. The k_J term listed for many of the entries indicates how the width depends on J [$k_p = k_0 + k_J J(J+1)$]. Except for the results by Eidelsberg et al. (1991), which were based on low-resolution spectra of absorption against continua from a synchrotron, the other values given in this Table are studies involving laser excitation. For the most part, there is general agreement among all results. Factors of a few difference (at most) exist between our widths and the order of magnitude evaluations given by Eidelsberg et al. (1991). Although the number of results for comparison with those from laser studies is smaller, the agreement is very good. The most significant differences are found with the widths derived by Drabbels et al. (1993), especially in light of the fact that the other studies yield a set of consistent results. The close correspondence with the laser studies, when available, gives us confidence in our other determinations. These will provide modelers of CO photodissociation with a more complete set of predissociation rates. We also note that since the rates have values greater than 10^{10} s^{-1} , the corresponding predissociation lifetimes are 0.1 ns or less, which are significantly less than radiative lifetimes ($\sim 1 \text{ ns}$). Thus, predissociation represents the primary decay channel for the upper states involved in the $B6$, $W0$, $W1$, $W2$, and $W3$ transitions studied here.

4. Concluding Remarks

We presented results for oscillator strengths and predissociation rates for Rydberg transitions in $^{12}\text{C}^{16}\text{O}$, $^{13}\text{C}^{16}\text{O}$, and $^{13}\text{C}^{18}\text{O}$. In particular, the $E-X$ (1-0), $B-X$ (6-0), and $W-X$ ($v' = 0, 1, 2, 3-v'' = 0$) bands were studied via absorption of synchrotron radiation. This suite of measurements represents the most complete database available at the present time for these transitions. Our oscillator strengths agree essentially with earlier determinations and expand upon available results for the rarer isotopologues. The same applies to the values

for predissociation rates, but now our larger set of measurements provides J -dependent rates for most of the transitions in the present study.

Our measurements are especially useful for chemical models that incorporate details of CO photodissociation. Oscillator strengths and predissociation rates are now available to examine selective isotope photodissociation with greater precision. We note that the significant line widths for the $B - X$ (6-0) and $W - X$ ($v' = 0, 1, 2, 3$ -0) bands minimize the importance of these transitions to selective self shielding, where the more abundant forms of CO are protected from dissociation, because optical depths at line center cannot reach large values.

With the tools now available to us, we plan to extend our set of measurements to other transitions below 1000 Å. We will utilize the high-resolution beam line on SOLEIL, the next-generation synchrotron in France, which is nearing completion with first light expected in 2007. High resolution is necessary to extract predissociation widths for the transitions important for modeling CO photodissociation in PDRs.

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Table 1. CO Oscillator Strengths $\times 10^3$

Band	λ (Å)	Isotopologue	Present 295 K	E91 ^a 295 K	C93 ^b 295 K	S91,92,93 ^c 295 K	Y95 ^d 20 K	Z97 ^e 295 K	S03 ^f ISM
<i>E1</i>	1051.71	1216	3.6 ± 0.3	2.5 ± 0.2	3.53 ± 0.35	3.0 ± 0.3	...	4.67 ± 0.66	3.3 ± 1.1
	1052.28	1316	4.2 ± 0.5	2.5 ± 0.2	...	3.1 ± 0.3
	1052.81	1318	3.5 ± 0.1	2.5 ± 0.2
<i>B6</i>	1002.56	1216	8.3 ± 0.7	7.9 ± 0.8	...	7.8 ± 0.8
	1002.81	1316	7.1 ± 0.4	7.3 ± 0.7
	1003.21	1318	5.0 ± 0.3	7.3 ± 0.7
<i>W0</i>	972.70	1216	16.6 ± 1.6	12.1 ± 1.2	...	12.9 ± 1.3	13.6 ± 2.0
	972.99	1316	15.1 ± 0.7	13.2 ± 1.3
	973.30	1318	13.8 ± 2.0	13.2 ± 1.3
<i>W1</i>	956.25	1216	16.0 ± 1.3	13.5 ± 1.4	...	14.8 ± 1.5	14.8 ± 1.5	...	15.8 ± 2.0
	956.29	1316	16.1 ± 2.8	16.1 ± 1.6
	956.33	1318	...	16.0 ± 1.6
<i>W2</i>	941.17	1216	30.8 ± 2.4	25.8 ± 2.6	...	30.0 ± 3.0	20.4 ± 3.1	...	23 ± 5
	941.65	1316	29.1 ± 1.3	27.9 ± 2.8
	942.16	1318	28.5 ± 2.0	27.9 ± 2.8
<i>W3</i>	925.81	1216	19.7 ± 1.4	16.3 ± 1.6	...	14.9 ± 1.5	17.0 ± 2.6	...	19.8 ± 2.4
	927.24	1316	18.7 ± 1.4	18.7 ± 1.9
	928.58	1318	15.4 ± 2.4	18.6 ± 1.9

^aEidelsberg et al. 1991.

^bChan et al. 1993.

^cStark et al. 1991, 1992, 1993.

^dYoshino et al. 1995.

^eZhong et al. 1997.

^fSheffer et al. 2003.

Table 2. Predissociation Rates (10^{11} s^{-1})

Band	Isotopologue	Parity	Present	E91 ^a	L92a ^b	L92b ^c	D93 ^d	E94 ^e	K95 ^f
<i>B6</i>	1216	e,f	13.0(1.0) ^{g, h}	3.3
	1316	e,f	2.3(0.1)	3.3
	1318	e,f	33.8(3.6)	3.3
<i>W0</i>	1216	e	0.12(0.01)+0.024(0.012) <i>x</i>	0.1	...	0.36(0.19)	0.070(0.004)+0.0264(0.0006) <i>x</i>	0.10(0.01)+0.022(0.002) <i>x</i>	...
		f	0.12(0.01)	0.096(0.008)
	1316	e	0.10+0.020 <i>x</i> ⁱ	0.1	...	≤0.3	...	≤0.3+0.013(0.002) <i>x</i>	...
		f	0.10 ⁱ
	1318	e	0.11(0.01)+0.01(0.01) <i>x</i>	0.1
		f
<i>W1</i>	1216	e,f	7.2 ⁱ	3.3
	1316	e,f	7.2 ⁱ	3.3
<i>W2</i>	1216	e	1.0(0.2)+0.034(0.013) <i>x</i>	1.0	1.15(0.15)	1.0(0.2)	3.86(0.21)	1.21(0.10)	≤1.9
		f	1.0(0.2)+0.018(0.002) <i>x</i>
	1316	e	0.76(0.21)+0.042(0.021) <i>x</i>	1.0	0.58(0.13)	0.47(0.19)	...	0.75(0.15)	...
		f	0.76(0.21)+0.021(0.010) <i>x</i>
	1318	e	0.42(0.06)+0.038(0.017) <i>x</i>	1.0
		f	0.42(0.06)+0.021(0.010) <i>x</i>
<i>W3</i>	1216	e	1.6(0.6)+0.13(0.02) <i>x</i>	3.3
		f	3.6(0.3)+0.040(0.004) <i>x</i>
	1316	e	1.0(0.1)+0.22(0.07) <i>x</i>	3.3
		f	3.1(0.7)+0.072(0.013) <i>x</i>
	1318	e,f	4.4 ⁱ	3.3

^aEidelsberg et al. 1991.

^bevelt et al. 1992a.

^cevelt et al. 1992b.

^dDrabbels et al. 1993.

^eEikema et al. 1994.

^fKomatsu et al. 1995.

^gWhen the predissociation rate is known to have a J dependence, the value is given by $k_p = k_0 + k_J x$, where x is $J(J+1)$.

^hUncertainties given in parentheses.

ⁱHeld fixed during the profile synthesis.

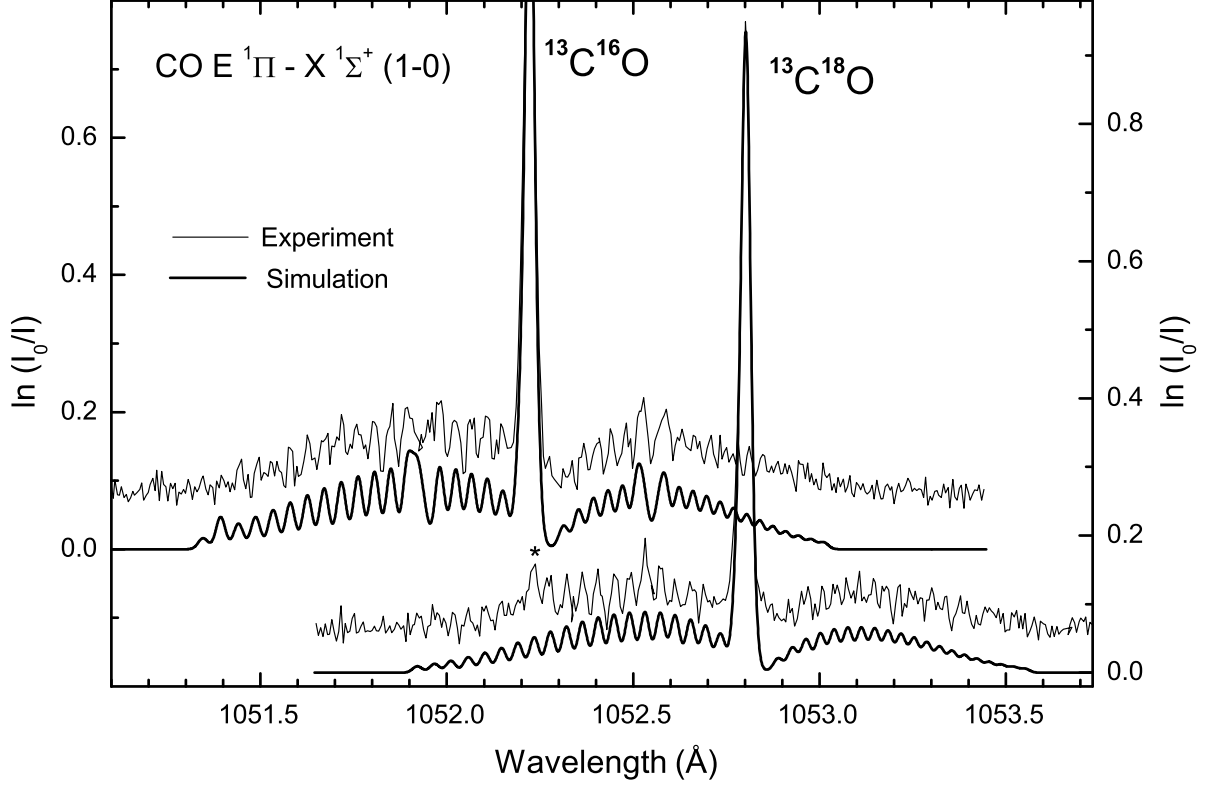


Fig. 1.— Experimental and best fitting simulated spectra of the $E1$ band for $^{13}\text{C}^{16}\text{O}$ and $^{13}\text{C}^{18}\text{O}$. The experimental spectra have been shifted vertically for clarity here and in Figures 2 and 3. The scale on the right hand side refers to the spectrum of $^{13}\text{C}^{18}\text{O}$. Note the local perturbations at $J = 7$ for $^{13}\text{C}^{16}\text{O}$. The feature marked by an asterisk is due to the impurity $^{13}\text{C}^{16}\text{O}$.

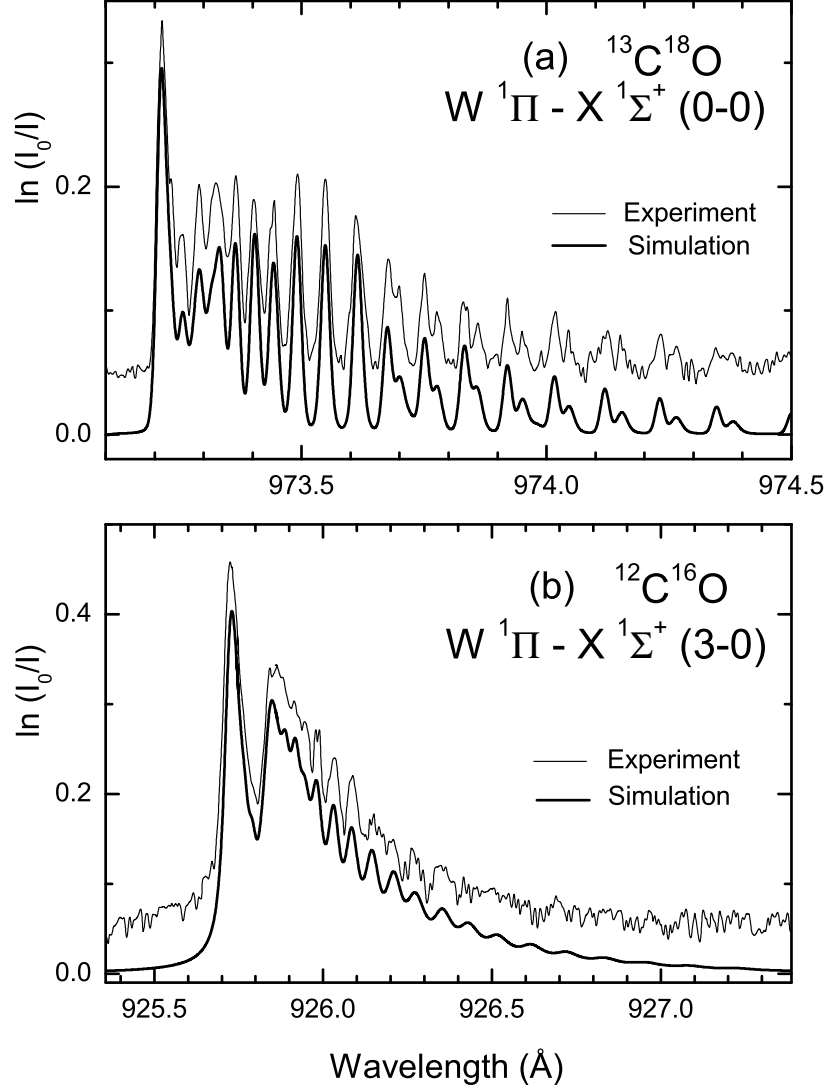


Fig. 2.— Experimental and best fitting simulated spectra of the $W0$ band for $^{13}\text{C}^{18}\text{O}$ (a) and the $W3$ band for $^{12}\text{C}^{16}\text{O}$ (b). The considerably broader lines in $W3$ reveal stronger predissociation (see data in Table 2).

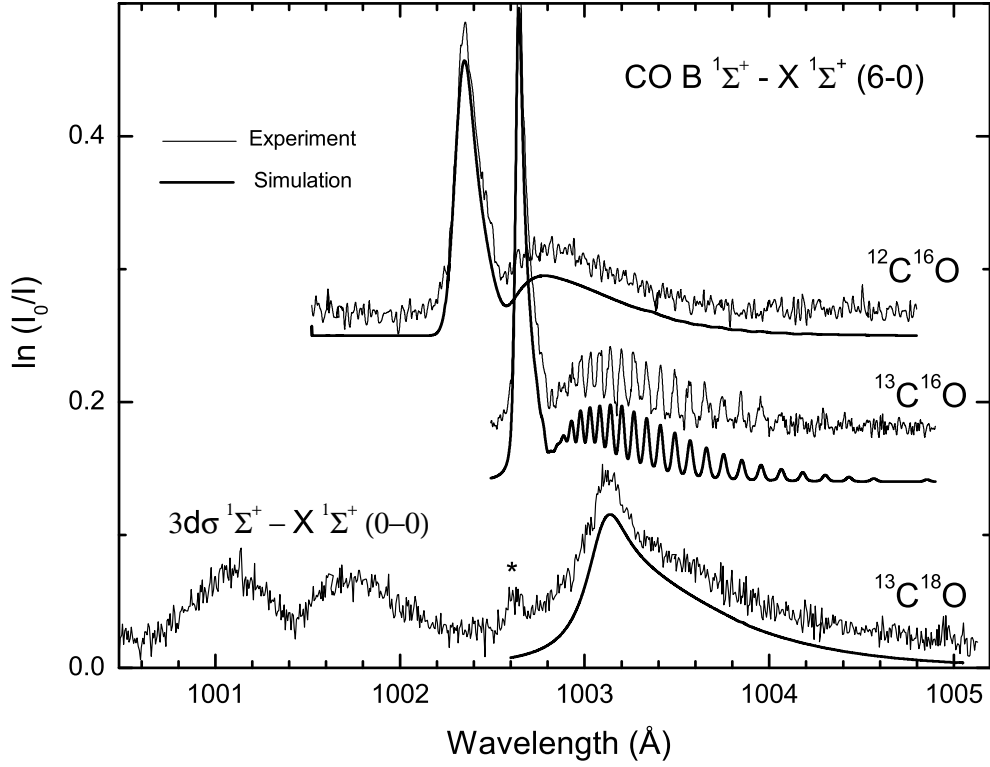


Fig. 3.— Experimental and best fitting simulated spectra of the $B6$ band for $^{12}\text{C}^{16}\text{O}$, $^{13}\text{C}^{16}\text{O}$ and $^{13}\text{C}^{18}\text{O}$. The spectra for $^{12}\text{C}^{16}\text{O}$ and $^{13}\text{C}^{16}\text{O}$ have been shifted upward for clarity. The feature marked by an asterisk is due to the impurity $^{13}\text{C}^{16}\text{O}$. The $3d\sigma$ forbidden band also appears in the $^{13}\text{C}^{18}\text{O}$ spectrum. For this isotopologue, the upper state is strongly mixed with $B6$ and borrows about half its intensity.